Investigation of the static magnetic properties of antiferromagnetic $CsMnF_3$ and $FeCl_2$

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(Submitted 12 May 1981)
Zh. Eksp. Teor. Fiz. **82**, 203–210 (January 1982)

The magnetic properties of single crystals of $CsMnF_3$ and $FeCl_2$ have been investigated in magnetic fields up to 60 kOe, at $T = 4.2$ K, with a vibrating-sample magnetometer that made it possible to measure three mutually perpendicular components of the magnetic moment. In $CsMnF_3$ there was detected a magnetic moment along the sixfold axis, occurring for definite positions of the antiferromagnetic vector L in the basal plane of the crystal. In $FeCl_2$, besides the magnetic moment along the C_3 axis observed and investigated earlier [A. N. Bazhan and V. A. Ul'yanov, Sov. Phys. JETP **52**, 94 (1980)], a magnetic moment was observed in the basal plane of the crystal, perpendicular to the applied magnetic field in this same plane.

PACS numbers: 75.30.Cr, 75.50.Ee

Antiferromagnetic $CsMnF_3$, of hexagonal symmetry D_{6h}^4, and $FeCl_2$, of rhombohedral symmetry D_{3d}^5, are among the materials that have been quite well studied.[1-8] In such hexagonal and rhombohedral crystals, as was shown in Turov's monograph,[3] weak ferromagnetism is possible along the six- or threefold axis, depending on the orientation of the vector **L** in the basal plane of the crystal. This ferromagnetic moment is caused by interaction of a form common to these two materials,

$$d[(\gamma_x+i\gamma_y)^3+(\gamma_x-i\gamma_y)^3]m_z$$

in the thermodynamic potentials that describe the magnetic properties of $CsMnF_3$ and $FeCl_2$.

The magnetic properties of $CsMnF_3$ were studied in Ref. 2. Below $T_N = 53.6$ K, single crystals of $CsMnF_3$ go over to an antiferromagnetic state, with the antiferromagnetic vector **L** oriented in the plane perpendicular to the hexagonal axis C_6. From a paper of Mil'ner and Popkov[4] on investigation of the magneto-optic properties of $CsMnF_3$, it follows that some of these properties can be explained by the occurrence of weak ferromagnetism along the sixfold axis.

The magnetic properties of single crystals of $FeCl_2$ were studied in Refs. 5–8. Below $T_N = 23.5$ K, $FeCl_2$ goes over to an antiferromagnetic state, with the antiferromagnetic vector **L** oriented along the threefold axis of the crystal. As was pointed out in Refs. 1, 3, and 6–8, it is of interest to study the phase transition of $FeCl_2$ when an applied magnetic field is oriented in the basal plane of the crystal. Furthermore,[1,6-8] for certain orientations of the magnetic field in the basal plane and for a certain value of the field, there can occur in $FeCl_2$ not only a magnetic moment along the applied magnetic field, but also one along the trigonal axis C_3 and one in the basal plane of the crystal, perpendicular to the applied magnetic field. The magnetic moment that occurs along the trigonal axis of an $FeCl_2$ crystal, for certain orientations of the magnetic field in the basal plane, was studied by us earlier.[1] The magnetic moment that occurs in the basal plane of the crystal, perpendicular to the applied magnetic field, $M_\perp(H)$, can exist also in $CsMnF_3$, if there is a component of the antiferromagnetic vector **L** along the hexagonal axis C_6.[3] But this magnetic moment is zero in $CsMnF_3$, since the antiferromagnetic vector **L** is perpendicular to the C_6 axis.[2]

The purpose of the present work was to observe and investigate directly the weak ferromagnetism along the sixth-order axis in $CsMnF_3$ and the magnetic moment in $FeCl_2$ that occurs in the basal plane of the crystal, perpendicular to the applied magnetic field. Investigation of the magnetic moments $M_\perp(H)$ perpendicular to the applied magnetic field in $CsMnF_3$ and $FeCl_2$ was carried out with vibrating-sample magnetometer that made it possible to measure three mutually perpendicular components of **M**.[9]

EXPERIMENTAL RESULTS

For description of the experiments done with the magnetometer, as in the preceding paper,[1] we choose a system of coordinates for the orientation of the magnetic field with respect to the crystal axes and the axes of measurement of the magnetic moment in the apparatus. Let the x, y, z axes be connected with the direction of measurement of the magnetic moment in the apparatus.[9] Then we denote by $M_i(H_j)$ the magnetic moment measured along direction $i(x, y, z)$ when the applied magnetic field H is oriented along direction $j(x, y, z)$.

The crystals were so mounted in the apparatus that their highest-order axes (C_6 for $CsMnF_3$ and C_3 for $FeCl_2$) were oriented along the vertical axis z of the apparatus. The magnetic field, in the (x, y, z) system, was oriented in the apparatus along the x axis. By rotation of the specimens about the z axis, we obtained a change of orientation of the applied magnetic field with respect to crystallographic directions, in the basal plane of the crystals.

For a single crystal of $CsMnF_3$, the variation $M_x(H_x)$ that we obtained for the magnetic moment measured along a magnetic field **H** applied in the basal plane of the crystal is linear and is described by the expression $M_x(H_x) = \chi_\perp H$, where $\chi_\perp = (4 \pm 0.1)\cdot 10^{-2}$ cgs emu/mol is the transverse magnetic susceptibility of $CsMnF_3$.

The value of the magnetic moment $M_x(H_x)$ is independent of the orientation of the magnetic field in the basal plane, and the numerical value of the magnetic suscepti-

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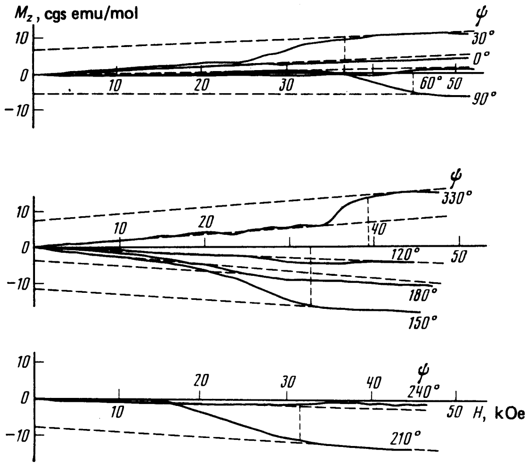

FIG. 1. Variation of the magnetic moment of $CsMnF_3$, measured along the hexagonal axis, with the applied magnetic field, for various orientations of the magnetic field in the basal plane.

bility agrees with the value of the magnetic susceptibility $\chi_\perp$ obtained in Ref. 2. Figure 1 shows the variation $M_z(H_x)$ of the magnetic moment of $CsMnF_3$ measured along the hexagonal axis of the crystal, for various orientations of the magnetic field in the basal plane. On change of the orientation of $\mathbf{H}$ in the basal plane of the crystal, there occurs a change both of the value of the magnetic moment $M_z(H_x)$ and of its sign.

Figures 2a and 2b show the variation $M_z(\psi)$ of the magnetic moment of the specimen with the angle ψ between the direction of the applied magnetic field, $H=50$ kOe, and the binary axis C_2, in the basal plane, for two successive mountings of the $CsMnF_3$ specimen in the apparatus. The difference between Fig. 2a and Fig. 2b consists in a different value of the angle α, the misalignment of the C_6 axis with the vertical axis z of the apparatus. As will be shown hereafter, because of the presence of domain structure in the magnetic moment along the sixfold axis, the value of the angle α plays an important role in the experiment.

The misorientation angle α is easily calculated by comparison of the slopes of the magnetization curves $M_z(H_x)$ at various angles ψ (Fig. 1), in magnetic fields $H<10$ kOe, and the slope of the magnetization curve $M_x(H_x)$ along the applied magnetic field. The $CsMnF_3$ crystal was mounted in the apparatus in such a way that the largest value of the angle α corresponded to orientation of the magnetic field, applied in the basal plane, along the binary axis of the crystal. It is evident from Fig. 1 that the magnetization curves $M_z(H_x)$ measured along the hexagonal axis C_6 of the crystal, in magnetic fields H larger than 30 kOe, can be described by the expression

$$M_z(H,\psi)=\sigma_z(\psi)+\chi^*(\psi)H.$$

The value of the spontaneous magnetic moment $\sigma_z(\psi)$ is determined by extrapolation of the linear magnetic-moment relation $M_z(H,\psi)$ in strong magnetic fields to $H=0$ (the dotted lines in Fig. 1). By determining the value of the magnetic moment $\sigma_z(\psi)$ for each aximuthal orientation of the field $\mathbf{H}\perp z$, one can plot the relation $\sigma_z(\psi)$, which is shown in Fig. 3. It agrees well with the $M_z(\psi)$ relation at $H=50$ kOe, shown in Fig. 2b.

To simplify the determination of the dependence $\sigma_z(\psi)$ of the spontaneous moment on the orientation of the applied magnetic field and to make the representation more graphic, the angle $\psi+120°$ in Figs. 2 and 3 corresponds to the angle ψ in Fig. 1. It is evident from Figs. 2b and 3 that the $\sigma_z(\psi)$ relation is described in the angular interval $0°<\psi<180°$ by the expression $\sigma_z(\psi)=\sigma_0|\sin 3\psi|$, where $\sigma_0=(6\pm 2)$ cgs emu/mol, and in the angular interval $180°<\psi<360°$ by the expression $\sigma_z(\psi)=-\sigma_0|\sin 3\psi|$. It must again be noted, however, that the $M_z(\psi)$ relations shown in Figs. 2b and 3 were obtained under the conditions indicated above regarding the angle α of misorientation of the axis C_3 and the axis z of the apparatus. The experimental arrangement described above is determined by the presence of a domain structure in the ferromagnetic moment σ_z in the crystal.

Figure 4 shows, for a specimen of $FeCl_2$, the variation $M_y(H_x)$ of the magnetic moment measured in the basal plane of the crystal, perpendicular to a magnetic

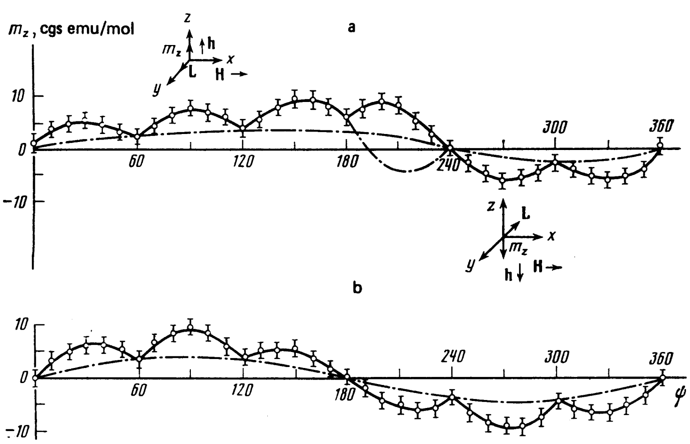

FIG. 2. Variation of the magnetic moment of $CsMnF_3$, measured along the hexagonal axis, with orientation of the applied magnetic field in the basal plane, for two mountings of the specimen in the apparatus; ψ is the angle between the direction of $\mathbf{H}$ and the axis C_2; $H=50$ kOe.

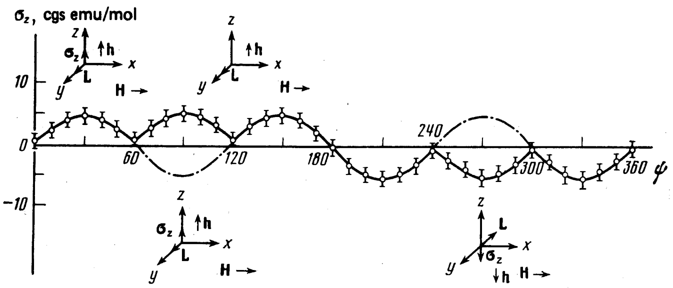

FIG. 3. Variation of the spontaneous magnetic moment σ_z along the axis C_6, in $CsMnF_3$, with the orientation of the applied magnetic fieldin the basal plane. Obtained by processing of the magnetization curves $M_z(H, \psi)$ of Fig. 1; ψ is the same as in Fig. 2.

field **H** applied in the same plane, for various orientations of the field with respect to the binary axis of the crystal. The angle ψ is the angle between the direction of the field **H** and the axis C_2 of the crystal. It is seen that for certain orientations of **H** in the basal plane of the crystal, there is a magnetic moment $M_y(H_x)$ perpendicular to **H** in this same plane, the value and sign of which depend on the orientation of **H**. When the magnetic field **H** is oriented at angle 30° to the binary axis, the variation $M_y(H_x)$ of the magnetic moment is described by the linear expression $M_y(H)=\chi^*H$. When the magnetic field is oriented along the binary axis, the variation is described by the nonlinear expression $M_y(H) = \chi_{I}H+\chi_{II}H^{\beta}$.

In order to determine the law that governs the variation of the value and sign of the magnetic moment $M_y(H_x, \psi)$ (Fig. 4) with the value and orientation of **H**, it is necessary to take into account, as was done earlier,[1] the contribution to the magnetic moment $M_y(H_x, \psi)$ from the component of the magnetic moment of the specimen in the basal plane, $M_{x'}(H_x)$, along the magnetic field. Such a contribution may arise because of inexact orientation of the crystal axes x', y', z' with respect to the measurement axes x, y, z of the magnetic moment in the apparatus. The correction for the magnetic moment $M_{x'}(H_x)$ is easily calculated by matching the $M_{x'}(H_x)$ relation and the linear section of the $M_y(H_x)$ relation. By processing the results shown in Fig. 4 in the same way as before,[1] one can plot the dependence of $M_y(H_0, \psi)$ on the orientation of the magnetic field in the basal plane. This dependence is shown in Fig. 5. It is seen that on change of the orientation of the magnetic field **H** in the basal plane of the crystal, the magnetic moment $M_y(H_x)$ changes according to the law

$$M_y(H, \psi)=M_y(H_0)\cos 3\psi+M_y^*(H_0)\sin\psi.$$

In order to determine the character of the nonlinearity

$$M_y(H)=\chi_{II}H^{\beta},$$

one can plot the variation of the value of log $M_y(H)$ with log H; but the accuracy of determination of the value of the parameter β as a result of such a plot, from our experiments, is poor. More accurate determination of this parameter requires experiments in stronger magnetic fields.

DISCUSSION OF RESULTS

According to Refs. 2 and 4, the antiferromagnetic vector **L** in $CsMnF_3$ is oriented in the basal plane of the crystal. On application of a magnetic field **H** in this plane, the antiferromagnetic vector **L** in the crystal can set itself perpendicular to **H** in two opposite, equivalent directions. Thus in the presence of weak ferromagnetism along the hexagonal axis C_6 (for certain orientations of **H** in the basal plane of the crystal),[3] a single crystal of $CsMnF_3$ may be split into domains, with the direction of σ_z along the axis C_6 in two opposite directions, connected with the directions of **L**. For exact orientation of the field in the basal plane of the crystal, because of the presence of a domain structure in σ_z along the axis C_6, we shall obtain no magnetic moment $M_z(H_x)$ along the measurement axis z. In order to detect and investigate the magnetic moment σ_z in $CsMnF_3$, it is necessary to apply a magnetic field **h** capable of changing the specimen to a single-domain state. For this purpose it is necessary to produce a misorientation of the vertical measurement axis z and of the axis

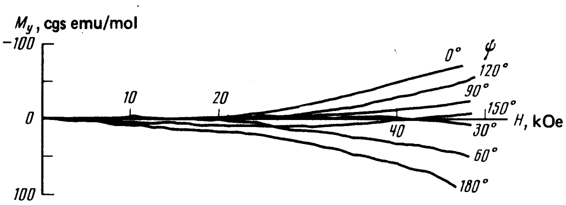

FIG. 4. Variation of the magnetic moment $M_y(H, \psi)$ of a specimen of $FeCl_2$, measured in the basal plane of the crystal and perpendicular to the applied magnetic field in the same plane, with the value of the field, for various orientations of H with respect to the binary axis of the crystal; ψ is the same as in Figs. 2 and 3.

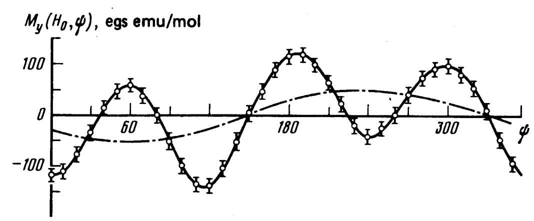

FIG. 5. Variation of the magnetic moment $M_y(H_0, \psi)$ of a specimen of $FeCl_2$ with orientation of the applied magnetic field in the basal plane (H_0 = 50 kOe). Obtained by processing of the magnetization curves $M_y(H, \psi)$ of Fig. 4.

C_6, in order that with increase of the magnetic field $\mathbf{H} \perp C_6$ there may appear along the axis C_6 a component $\mathbf{h}$ that reverses the specimen magnetization.

According to Turov,[3] the maximum value of the misorientation angle α should occur for orientation of $\mathbf{H}$ in the basal plane of the crystal along its binary axis. Then the antiferromagnetic vector $\mathbf{L}$ is oriented along the vertical plane of symmetry, and a state with weak ferromagnetism along the axis C_6 is possible. The experiment represented in Figs. 1 and 2 was conducted in this way. The maximum value of the angle α amounts to $\alpha \approx 1°$. By comparing the slopes of the magnetization curves $M_z(H, \psi)$ in Fig. 1 in strong magnetic fields, $H > 40$ kOe, with the slope of the $M_x(H_x)$ relation, one can determine the angle α between the axes z and C_6 and plot the $\sigma_z(\psi)$ relation. The results of this processing are shown in Fig. 3. Also shown there schematically are the relative orientations of the magnetic field $\mathbf{H}$, the magnetization-reversing field $\mathbf{h} \parallel C_6$, the antiferromagnetic vector $\mathbf{L}$, and the ferromagnetic moment σ_z.

The fact that the possibility of observation of the magnetic moment $M_z(H) = \sigma_z$ is determined by magnetization reversal of domains in the $CsMnF_3$ specimen is substantiated by the experiment represented in Figs. 2a and 2b. It is seen from Fig. 2a that the $M_z(\psi)$ relation can be described by the expression

$$M_z(\psi) = \sigma_z |\sin 3\psi| + F_I(\psi)$$

in the angular interval $0° < \psi < 240°$, whereas in Fig. 2b the $M_z(\psi)$ relation is described by the expression

$$M_z(\psi) = \sigma_z |\sin 3\psi| + F_{II}(\psi)$$

in the angular interval $0° < \psi < 180°$. The difference between the experiments of Figs. 2a and 2b, as has already been pointed out, consists in a different value of the angle α of misorientation of the axis z and the axis C_6.

The functions $F_I(\psi)$ and $F_{II}(\psi)$, represented in Figs. 2a and 2b by the dotted lines, are determined by the projection of the magnetic moment $M_x(H_x)$ in the basal plane of the crystal on the hexagonal axis. This projection arises because of the misorientation of the vertical measurement axis z and the axis C_6. The signs of $F_I(\psi)$ and $F_{II}(\psi)$ determine the sign of the magnetization-reversing field $\mathbf{h}$ along the axis C_6 in the crystal. For ideal rotation of the specimen, without deviation from the vertical axis, the relation $F(\psi)$ is determined by the expression

$$F(\psi) = F_{II}(\psi) = A \sin \psi.$$

As seen from Fig. 2b, the magnetization-reversing field $\mathbf{h} \parallel C_6$ changes here the specimen to a single-domain state with $\boldsymbol{\sigma}_z \parallel \mathbf{h}$ in the angular interval $0° < \psi < 180°$ and with $(-\boldsymbol{\sigma}_z) \parallel \mathbf{h}$ in the angular interval $180° < \psi < 360°$. For rotation of the specimen with deviation from the vertical measurement axis, a more complicated $F_I(\psi)$ relation occurs, Fig. 2a. Then orientation of the magnetization-reversing field $\mathbf{h}$ along the axis C_6 in one direction and a single-domain state of the specimen with $\boldsymbol{\sigma}_z \parallel \mathbf{h}$ were maintained within the angular interval $0° < \psi < 240°$. In the angular interval $240° < \psi < 360°$, the sign of the magnetization-reversing field changed, and accordingly so did the sign of the ferromagnetic moment σ_z. In the experiment, deviation of the specimen from the vertical axis during rotation was observed visually.

For theoretical interpretation of the data on the single crystal of $CsMnF_3$, it is necessary to write the thermodynamic potential that describes the magnetic properties of this material. It is known that the six magnetic Mn^{++} ions in an elementary cell of $CsMnF_3$ are located in two crystallographically nonequivalent positions, and that its properties are described by a model with six magnetic sublattices.[10] The thermodynamic potential of $CsMnF_3$ must be written on the basis of this model. But for a qualitative description of the observed phenomenon, we shall limit ourselves to the form of the thermodynamic potential of an easy-plane antiferromagnet, of hexagonal symmetry, with two ions per unit cell.[3] The thermodynamic potential of crystals of hexagonal symmetry D_{6h}^4 has the form

$$\Phi = {}^1/_2 B\mathbf{m}^2 + {}^1/_2 D(\gamma\mathbf{m})^2 + {}^1/_2 a\gamma_z^2 - {}^1/_2 d[(\gamma_x + i\gamma_y)^3 + (\gamma_x - i\gamma_y)^3] m_z - \mathbf{mH}, \quad (1)$$

where $\mathbf{m}$ is the magnetic vector, and where $\gamma = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$ is the unit antiferromagnetic vector.

On minimizing (1) with respect to m_x, m_y, and m_z and introducing the angle φ between the axis C_2 and the projection of the antiferromagnetic vector $\mathbf{L}$ on the basal plane, one finds that the magnetic moment along the sixfold axis is described by the expression

$$m_z = \frac{d}{B}[(\gamma_x + i\gamma_y)^3 + (\gamma_x - i\gamma_y)^3] = \frac{d}{B}\cos 3\varphi. \quad (2)$$

In Fig. 3, the dashed curve represents this relation. Thus on change of orientation of the antiferromagnetic vector $\mathbf{L} \perp \mathbf{H}$ in the basal plane of a $CsMnF_3$ crystal, a weak ferromagnetism σ_z along the hexagonal axis occurs, whose variation with the orientation of $\mathbf{H}$ is determined by the expression (2). Knowing the value $\sigma_0 = (6 \pm 2)$ cgs emu/mol, we can determine the value of the effective field $H_e = \sigma_z/\chi_\perp$ responsible for its origin: $H_e = (0.2 \pm 0.5)$ kOe.

We turn now to discussion of the results obtained on the single crystal of $FeCl_2$. As a result of experiments on the single crystal of $FeCl_2$ in a magnetic field $\mathbf{H}$ oriented in the basal plane, one can draw the conclusion that for certain orientations of $\mathbf{H}$, there arise magnetic moments $M_z(H_x, \psi)$ and $M_y(H_x, \psi)$ along the threefold axis C_3 and in the direction perpendicular to $\mathbf{H}$ in the basal plane. The dependence of the magnetic moments $M_z(H_x, \psi)$ and $M_y(H_x, \psi)$ once definite orientation of the field $\mathbf{H}$, is determined by the expression

$$M_y(H_0\psi) = M_y(H_0)\cos 3\psi, \qquad M_z(H_0\psi) = M_z(H_0)\sin 3\psi.$$

A theoretical interpretation of the results can be made on the basis of Ref. 6, with allowance for magnetoelastic interactions in the Hamiltonian that describes the magnetic properties of $FeCl_2$, and on the basis of Turov's book,[3] by investigation of the thermodynamic potential of crystals of symmetry D_{3d}^5, we considered a phase transition caused by rotation of the antiferromagnetic vector $\mathbf{L}$ from a state with $\mathbf{L} \parallel C_3$ to a state with $\mathbf{L} \perp C_3$ on increase of a magnetic field $\mathbf{H} \perp C_3$. Such a phase transition is possible[11,12] if the symmetry of the crystal allows the possibility of existence of a weak ferromagnetism in the basal plane of the crystal, and if its value is sufficient for production of such an orientational phase transition in a perpendicular field. In papers of Nasser and Varret,[7,8] it is stated that without allowance for magnetoelastic interactions, the rotation of the magnetic moments of the sublattices of $FeCl_2$ occurs in the plane passing through the applied magnetic field and the axis C_3. To determine how the magnetic moments of the sublattices in $FeCl_2$ rotate with increase of a magnetic field in the basal plane, neutron-diffraction experiments are necessary. Furthermore, as was shown in a paper of Nasser,[13] neutron-diffraction experiments with application of pressure would make it possible to explain the role of magnetoelastic interactions in $FeCl_2$.

Thus it can be stated that in $FeCl_2$, with a magnetic field oriented in the basal plane of the crystal, in addition to the magnetic moment $M_x(H_x)$ along the applied magnetic field **H**, there are magnetic moments $M_z(H_x\psi)$ along the axis C_3 and $M_y(H_x\psi)$ in the basal plane, perpendicular to **H** and having a 120-degree periodicity on change of the orientation of **H** in the basal plane of the crystal.

The magnetic moments along the sixfold axis in $CsMnF_3$ and along the threefold axis in $FeCl_2$ are determined by the projection of the antiferromagnetic vector **L** on the basal plane. In $CsMnF_3$, the antiferromagnetic vector **L** is oriented in the basal plane, and the quantity σ_z has its maximum value. In $FeCl_2$, in the absence of a magnetic field, the antiferromagnetic vector **L** is oriented along the trigonal axis. On increase of a magnetic field in the basal plane, the orientational phase transition gives rise to a projection of **L** on the basal plane and to magnetic moments $M_\perp(H_x)$ and $M_z(H_x)$ that depend on the applied magnetic field **H**.

The authors deeply and sincerely thank P. L. Kapitza for his interest in the research, A. S. Borovik-Romanov for direction of the research, and I. E. Dzyaloshinskii, N. M. Kreines, and V. A. Trunov for discussion of the results.

Note added in proof (1981, December 25): The curves in Figs. 4 and 5 and the discussion of the results for $FeCl_2$ allow for the splitting of the $FeCl_2$ specimen into domains with respect to the magnetic moment $M_y(H_x)$, when there is exact orientation of the applied magnetic field $H\|C_2$, and they allow for the change of sign of $M_y(H_x)$ during remagnetization of the specimen near the axis C_2.

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Translated by W. F. Brown, Jr.